Suggy's Research

Present and Future Research

My main research focus is on theoretical understanding of the dynamics of optical excitations, electrons, and charge carriers in macromolecules and nano-structured materials. Systems of concern include light harvesting complexes of purple bacteria, dendrimers, conjugated polymers, nano-wires based on organometallic compounds, DNA, and quantum dots. Two complementary approaches, one directly addressing issues pertaining to new experimental measurements and the other implementing molecular level simulations, are being performed for the following purposes: (i) to understand the fundamental molecular processes responsible for important optical and conductance properties; (ii) to provide important chemical information helpful for building up better nano-scale energy and information processing devices.

Research on bacterial light harvesting complexes is currently in progress. A new theoretical formalism was developed to this end, which addresses important issues regarding modern spectroscopic experiments of macromolecules in general. I plan to adapt this theory for the understanding of single molecule and nonlinear spectroscopies of dendrimers, conjugated polymers, DNA, and quantum dots, and develop the theory further for the understanding of conductance measurements on nano-wires and DNA. In addition, I plan to implement modeling and simulation, especially, of organo-metallic nanowire, DNA, dendrimers, and conjugated polymers so as to gain better microscopic understandings of these systems. Various methods will be employed including nonequilibrium molecular dynamics, path integral simulation, and quantum master equation approaches, and the design of simulation will always be made such that experimental verifications are possible.

Past Research

- **Open system quantum dynamics**

  A fourth order quantum master equation was derived for a general system Hamiltonian. This equation goes beyond the assumption of weak system-bath interaction implicit in the Redfield equation, and can be employed for the study of the effects of moderately large environmental coupling on the dynamics of nanoscale and biological systems.

- **Quantum control**

  A general theory for the optimal quantum control in dissipative environments was developed which accounts for the interplay of optical fields and nonMarkovian bath relaxation in a general manner. The theory will enable to understand a systematic way to manipulate the laser pulse in order to exploit the environmental perturbation in a constructive manner.

- **Energy transfer reaction**

  The Förster-Dexter theory for the excitation energy transfer reaction was generalized for the nonequilibrium situation where some of the bath modes relax in time scales comparable to that of the energy transfer reaction. The theory relates the reaction rate to an ultrafast pump-probe experiment, providing means for the confirmation of the theory and for experimental determination of the reaction parameters.

- **Electron transfer reaction**

  For multistate electron transfer reaction systems with a quantum reaction coordinate in the deep tunneling regime, electronic interference effects were studied employing a nonadiabatic generalization of the instanton theory. A large interference effect was observed for the cases with multiple elctronic channels.

- **Photosynthesis**

  A general statistical analysis was made on a model of the B850 band of the light harvesting complex 2 (LH2), a peripheral light harvesting unit found in purple bacteria.

Universal relations among the level spacings and the inhomogeneities of optically important excitonic states were found, which can be used for experimental verifications of the structural nature of the system and characteristics of the disorder.

- **Path integral theories**

  A rigorous formulation was developed for the real time dynamics of the path integral centroid, which was then used for a theoretical justification of the centroid molecular dynamics (CMD), an approximate and practicable condensed phase simulation method that had lacked a firm theoretical basis. The new formulation enabled extensions and improvements of the CMD method in various directions.

- **Path integral simulation methods**

  A higher order composite factorization method for the imaginary time path integral simulation was developed. This method is more efficient that that based on the primitive factorization by about a factor of four and can be used for the CMD simulation. Numerical tests show that the new method is advantageous for systems such as water and an excess electron in liquid.

- **Path integral quantum rate theory**

  A general reaction rate theory for quantum activated processes was developed based on the path integral centroid dynamics perspective. The theory clarified the relationship between the CMD method and the path integral quantum transition state theory, and provided an improvement of the latter by accounting for the anharmonic contribution of the quantum transmission factor. The theory can be implemented for a general multidimensional system and cover both the deep quantum tunneling regime and the classical limit.

- **Simulation of quantum reaction dynamics**

  An application of the path integral quantum transition state theory was made for the calculation of the recombination rate of lithium atoms in solid hydrogen. Simulation techniques such as constraint molecular dynamics and constant pressure path integral simulation were used, and experiences on these techniques will be of great value for the implementation of the new quantum reaction rate theory for realistic systems.

- **Diffusion limited reaction rate theory**

  The interplay of the translation diffusion and the long range energy transfer reaction in the transport of excitation in solution phase was studied employing a manybody diffusion limited reaction rate theory. Enhancement of the excitation migration with the increase of the mobility of the chromophore was quantified in various limits, which are consistent with experimental observations.